NH_3 inverse modeling results using new TES NH_3 observations, and surface measurements.

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Abstract:

Emissions of ammonia pose a concern to the environment for several reasons. Ammonium nitrate and ammonium sulfate make up a substantial fraction of atmospheric fine particulate matter ($PM_{2.5}$), exposure to which will cause health problems. Further, when deposited in excess, reactive nitrogen, including ammonia, can cause detrimental nutrient imbalances to sensitive ecosystems. However, there are lots of uncertainties in ammonia emission inventories. The uncertainty is varied and can be the total amount of emissions or even the daily variations. Our goal is to constrain ammonia emissions estimates using adjoint methods (GEOS-Chem) and new measurements. To achieve this goal, we first explore the capabilities and limitations of the inverse modeling framework system. We begin with a forward model simulation of atmospheric NH_3 concentrations given a known NH_3 emission inventory. The TES radiative transfer model is applied to this atmospheric state, and pseudo TES NH_3 retrievals are generated which have sampling and error characteristics of actual TES NH_3 observations. Performing inverse modeling with pseudo data allows us to quantify the bias of the inversion. Inversions using real data are then performed for several months in 2008. We evaluate the inverse modeling results by comparing the observationally constrained model simulations to independent surface measurements of NH_3 (AMoN) .

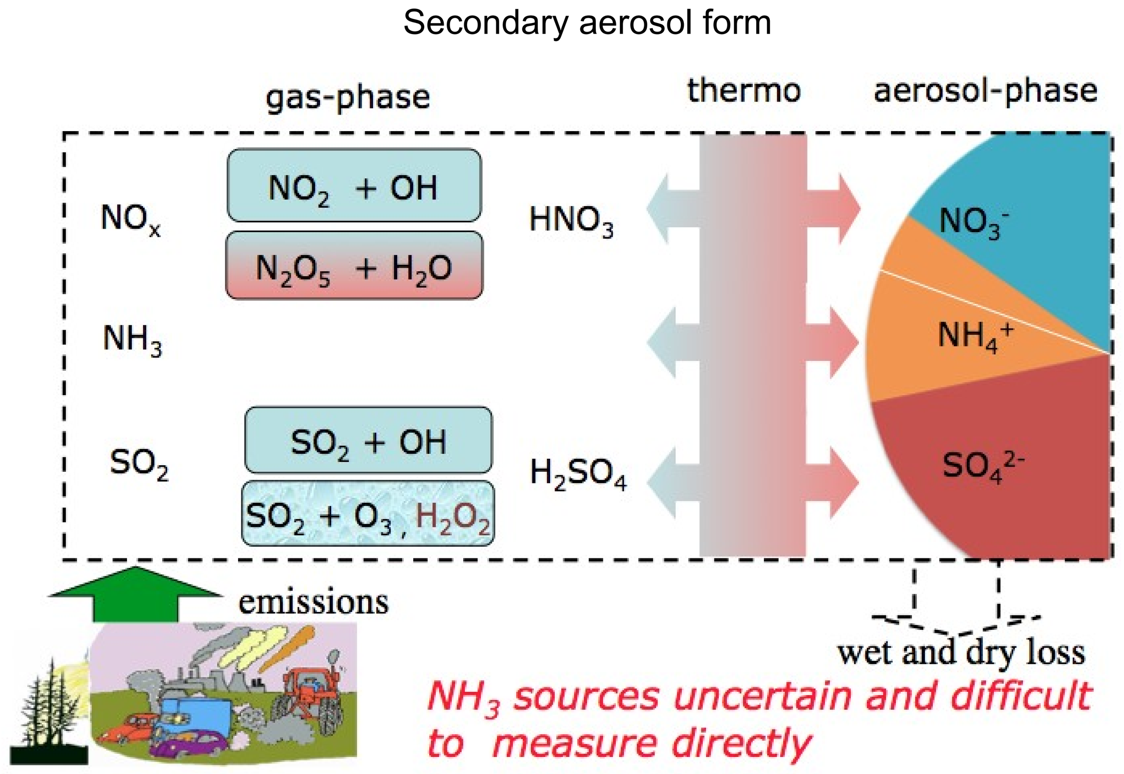
Why study NH_3 ?

– A substantial fraction of $PM_{2.5}$. Health impacts

– Importance for $PM_{2.5}$ control

– Affect climate change

Secondary aerosol form

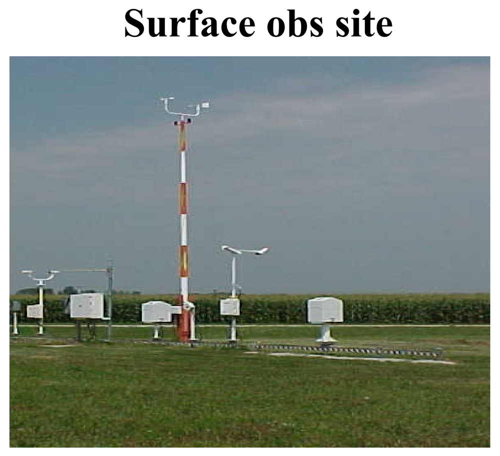


NH_3 sources uncertain and difficult to measure directly

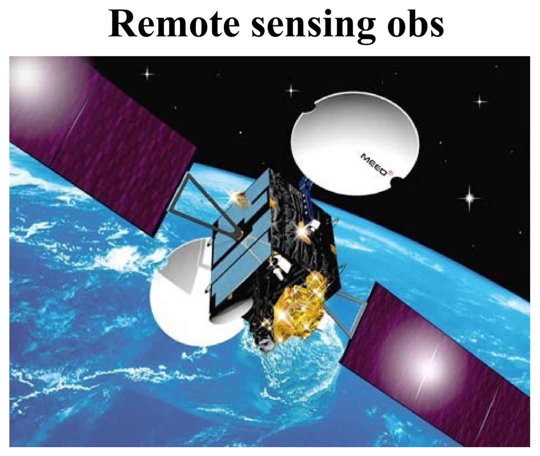
Objectives:

Constrain NH_3 emission using new remote sensing observation (TES NH_3 retrievals), existing surface observation (AMoN), and GEOS-Chem adjoint modeling tools.

Surface obs site

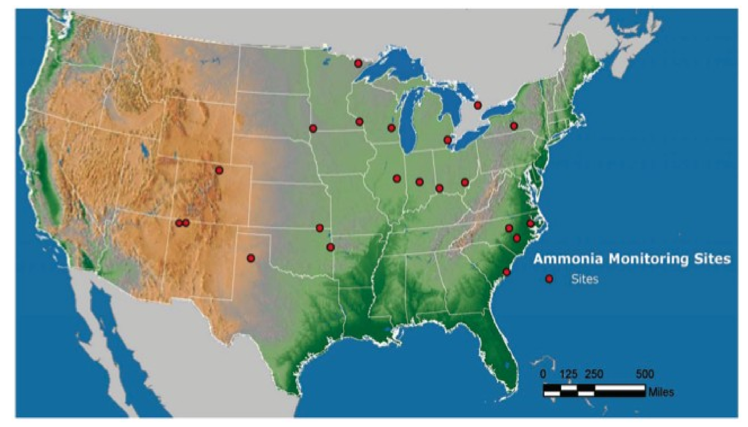


Remote sensing obs



AMoN surface obs: 21 sites in the U.S.

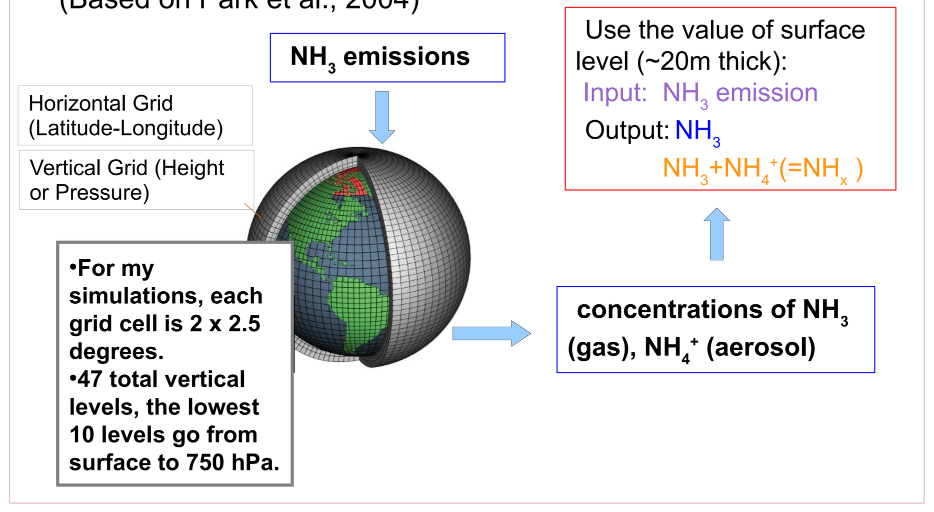
Ammonia Monitoring Network



AMoN sites (collected by *Gary Lear*)
http://www.epa.gov/

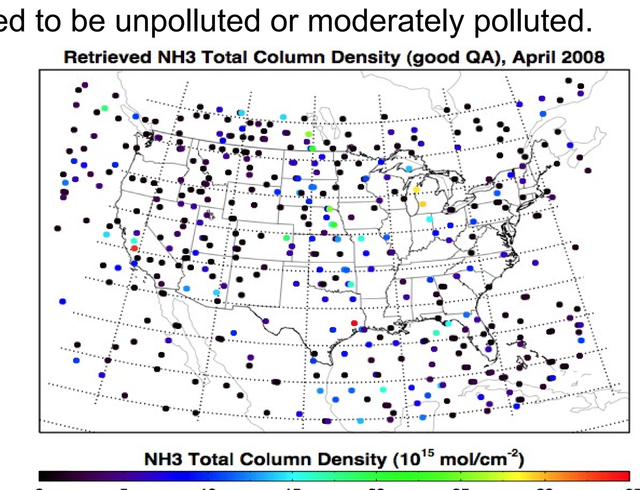
GEOS-Chem model, v8-02-03

(Based on Park et al., 2004)

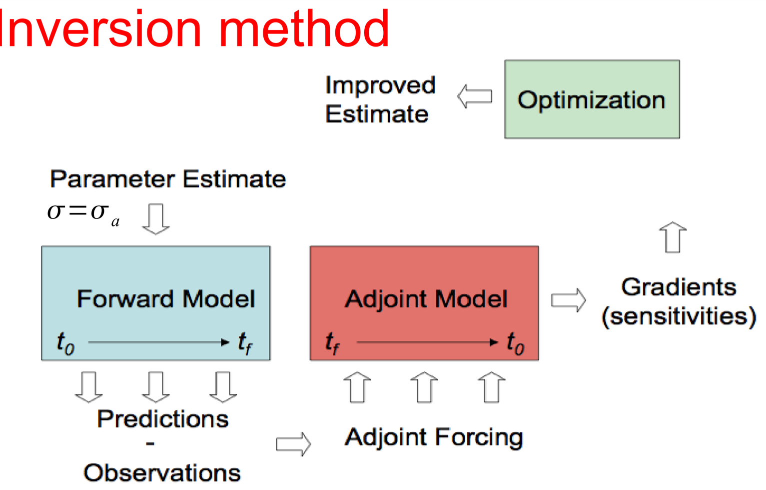


TES: remote sensing of NH_3

- There are three types of a priori NH_3 profiles:
  – Unpolluted: NH_3 < 1ppbv
  – Moderately Polluted: 1< NH_3 < 5ppbv (below 500 mb)
  – Polluted: NH_3 > 5ppbv (surface)
- For the following retrievals, a priori NH_3 profiles are selected to be unpolluted or moderately polluted.



Inversion method

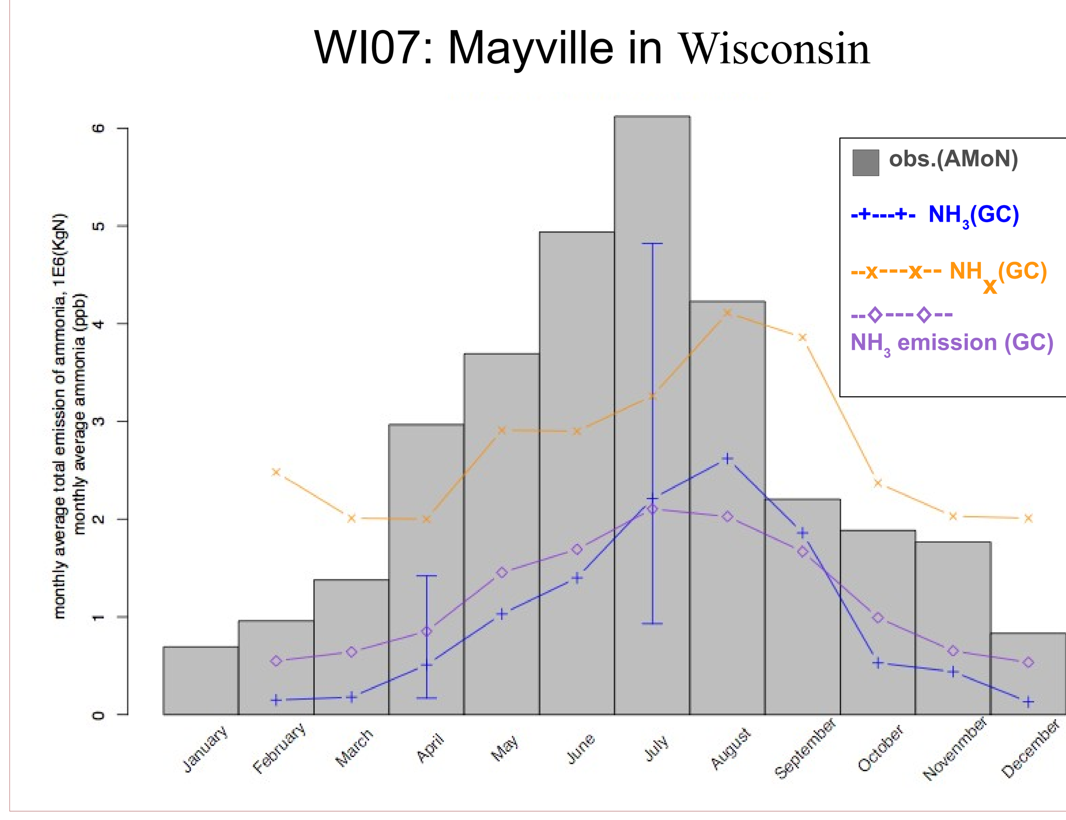


– GEOS-Chem adjoint (Henze et al., 2007, 2009)
– Cost Function J (want to minimize):

$$J=\frac{1}{2}\sum_{c\in\Omega}(Hc-\boldsymbol{c}_{obs})^T\boldsymbol{S}_{obs}^{-1}(Hc-\boldsymbol{c}_{obs})+\frac{1}{2}\gamma_r(\sigma-\sigma_a)^T\boldsymbol{S}_a^{-1}(\sigma-\sigma_a)$$

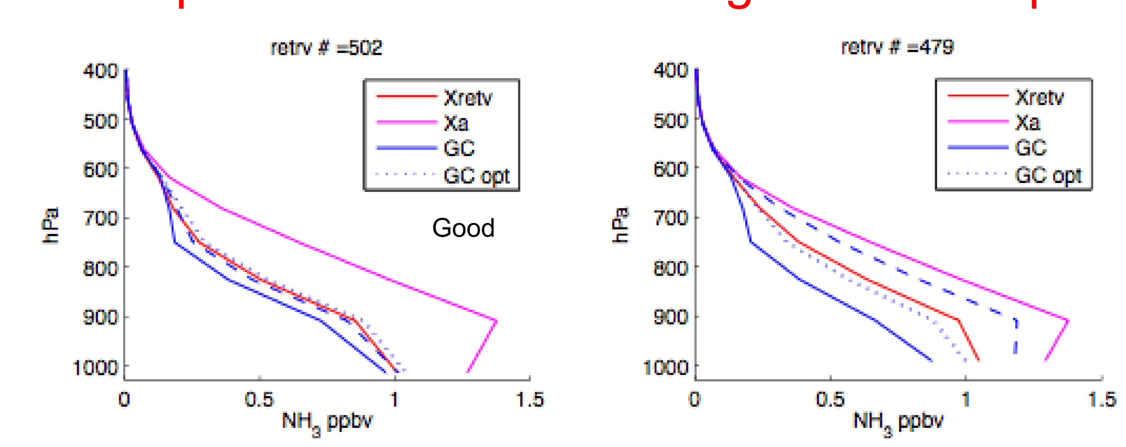
– H = TES obs operator
– c = Model NH3 profile
– c_{obs}= TES NH3 profile
– S = Error covariance matrices
– γ_r= Regularization parameter
– σ = Emissions scaling factors for each grid, $\sigma=\ln(\frac{emission}{emission_a})$
– σ_a= Initial guess of scaling factos (=0)

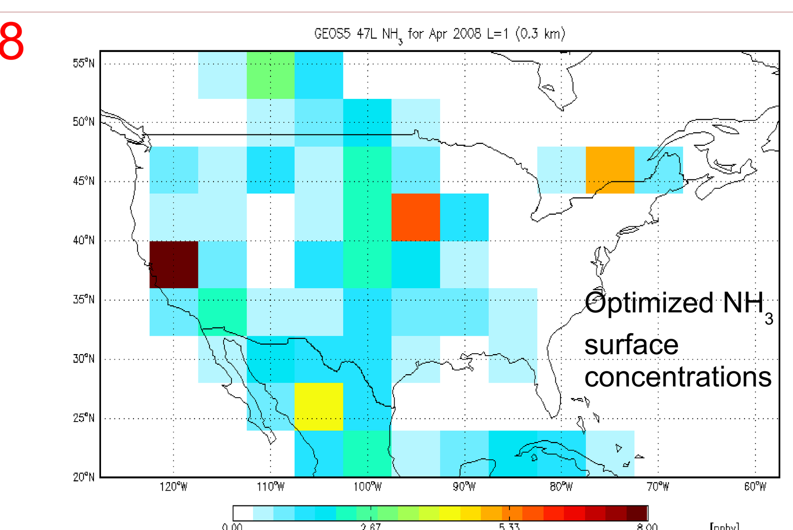
Initial comparison of GEOS-Chem model and AMoN obs: NH_3 (ppb)



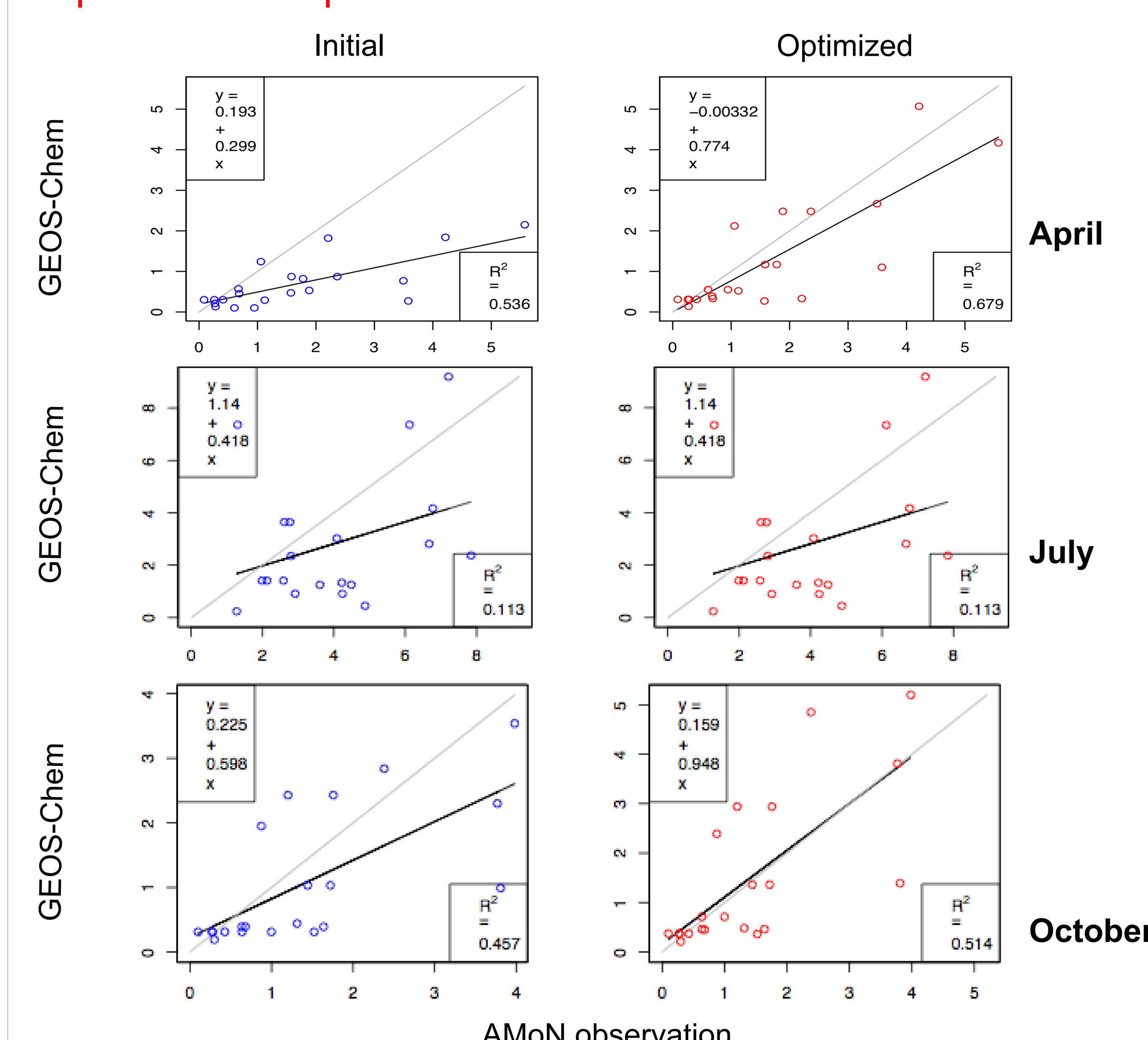
- Seasonality of GEOS-Chem correlated to AMoN obs
- Magnitude of GEOS-Chem and AMoN do not match. Reasons may be:
  ►GC: NH_3 emissions constant within each month *(has seasonal variation, but constant during one month)*;
  AMoN: real data, influenced by day-to-day variability.
  ►GC: 2005; AMoN: 2007~2009. *(although AMoN values do not show significant inter-annual variability)*
  ►GC: emissions may not be right.
  ►GC: deposition may not be right.
  ►GC: average of a 2°*2.5° region;
  AMoN: point measurement.

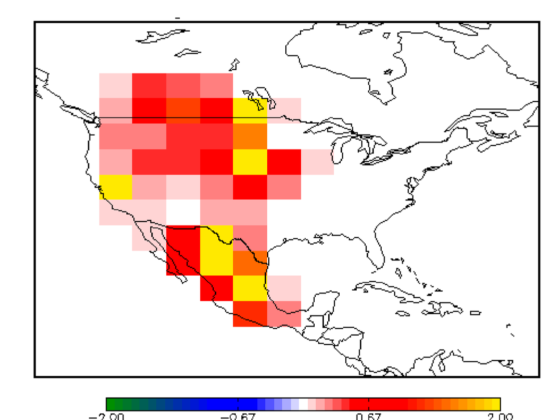
The optimized emissions using TES obs: April 2008





Optimized comparison of GEOS-Chem model and AMoN obs: NH_3 (ppb), 2008





$\sigma=\ln(\frac{emission_{optimized}}{emission_{initial}})$

– For July, we have only finished 5 iterations. So it didn't improved much from the initial comparison.

– For April and October, the optimized GEOS-Chem model results are better correlated and have less bias in comparison to AMoN.

– We are continuing to develop and carry out these inversions as well as analyzing data from additional months.

Reference:
1. Beer, R., M. W. Shephard, S. Kulawik, S. A. Clough, R. A. Eldred, K. W. Bowman, S. P. Sander, B. M. Fisher, V. H. Payne, M. Luo, G. B. Osterman, and J. R. Worden.: First satellite observations of lower tropospheric ammonia and methanol, Geophys. Res. Lett., 35, L09801, doi:10.1029/2008GL033642. 2008
2. Henze, D. K., Hakami, A., and Seinfeld, J. H.: Development of the adjoint of GEOS-Chem, Atmos. Chem. Phys., 7, 2413-2433, 2007.
3. Henze, D. K., Seinfeld, J. H. and Shindell, D. T.: Development of the adjoint of GEOS-Chem, Atmos. Chem. Phys., 9, 5877-2009.

Acknowledgement : **This research was support by NASA grants NNX09AN77G and NNX10AG63G as well as US EPA grant RD834559.**